FRACTAL CONCEPTS IN SURFACE GROWTH

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PART I Introduction

1 Interfaces in nature

Most of our life takes place on the surface of something. Sitting on a rock means contact with its surface. We all walk on the surface of the Earth and most of us don't care that the center of the Earth is molten. Even when we care about the interior, we cannot reach it without first crossing a surface. For a biological cell, the surface membrane acts not only as a highly selective barrier, but many important processes take place directly on the surface itself.

We become accustomed to the shapes of the interfaces we encounter, so it can be surprising that their morphologies can appear to be quite different depending on the scale with which we observe them. For example, an astronaut in space sees Earth as a smooth ball. However Earth appears to be anything but smooth when climbing a mountain, as we encounter a seemingly endless hierarchy of ups and downs along our way.

We can already draw one conclusion: surfaces can be smooth, such as the Himalayas viewed from space, but the same surface can also be rough, such as the same mountains viewed from earth. In general the *morphology depends on the length scale of observation*!

How can we describe the morphology of something that is smooth to the eye, but rough under a microscope? This is one question we shall try to answer in this book. To this end, we will develop methods to characterize quantitatively the morphology of an arbitrary interface. In fact, we shall see that concepts like roughness are replaced by exponents that refer not to the roughness itself, but to the fashion in which the roughness *changes* when the observation scale itself changes.

Fractal objects in nature are the same on different observation scales. However, the above examples look rather different when the scale is changed. Does this fact imply that the title of this book is a misnomer? Yes and no. In fact, many interfaces and surfaces are examples of *self-affine* objects, which are 'intermediate' between fractal objects and non-fractal objects in the following sense. When we make a scale change that is the *same in all directions*, self-affine objects change morphology. On the other hand, when we make a scale change that is *different for each direction*, then interfaces do not change morphology. Rather, they behave like fractal objects in that they appear the same before and after the transformation (see Fig. 1.1).

This book will explore in some detail the nature of such self-affine objects. We shall see that this feature is analogous to a 'symmetry principle.' Symmetry principles codified in group theory enable one

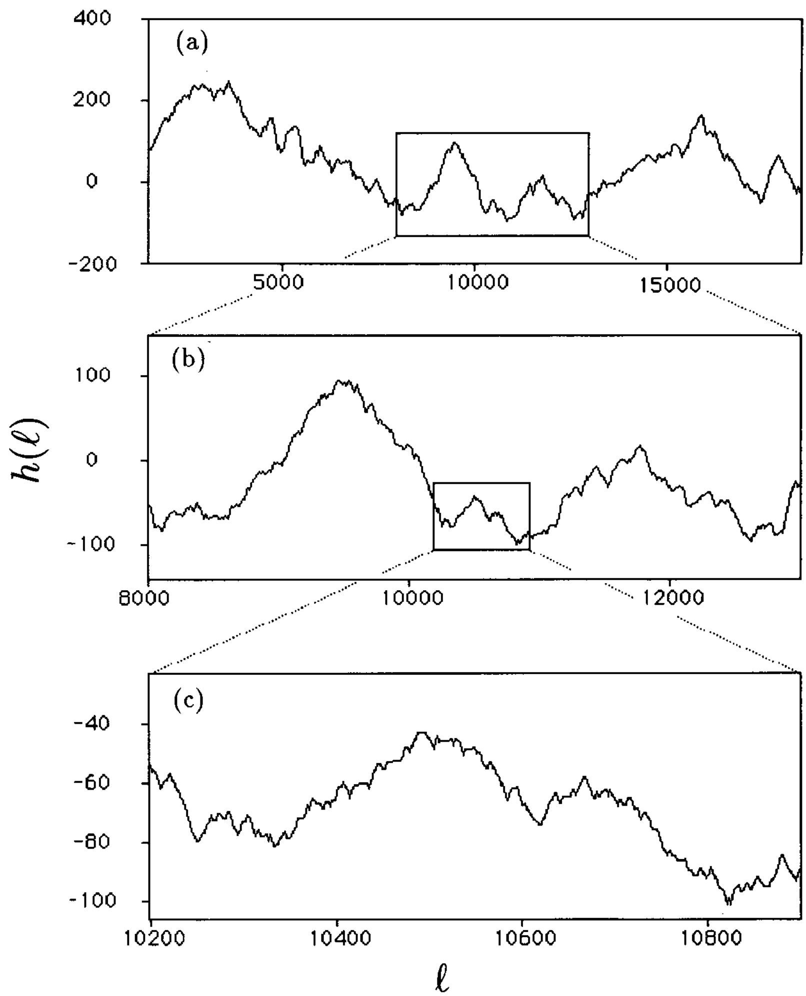


Figure 1.1 Rescaling a self-affine function, in this case the 'DNA walk' introduced in §1.3.2 (cf. Fig. 1.13). Only if the two unequal magnification factors, M_ℓ and M_h, by which the ℓ and h directions are re-scaled, are selected correctly will the enlarged portion have the same statistical properties as the original. (After [416].)

to classify and eventually understand some properties of a crystalline system. Similarly, the symmetry obeyed by self-affine objects will enable us to classify and perhaps better understand some properties of rough interfaces in nature.

We are interested not only in the *morphology* of various 'pre-formed' interfaces, but also in the dynamics of how the morphology develops in time. Some surfaces are formed as a result of a deposition process. Others shrink due to erosion or etching. Some interfaces propagate through inhomogeneous media. An interesting set of questions concerns the *formation, growth, and dynamics* of such interfaces.

In this first chapter, we offer the interested reader an *apéritif* – by exposing a variety of the themes of this book without the baggage imposed by requiring equations or discussing experimental details. The reader whose appetite is already 'up' for the main course is invited to proceed directly to Chapter 2.

1.1 Interface motion in disordered media

1.1.1 Fluid flow in porous media

A familiar scene: at breakfast, your spilled coffee suddenly conquers a segment of the tablecloth. Probably this is not the right moment to contemplate the microscopic forces that balance just when the coffee stops spreading. Nor is it the appropriate moment to start thinking about the shape of the patch, or the roughness of its surface. So wait until the stress subsides, and then examine the large brown patch.

By clipping a paper towel on a stand and immersing one end into a fluid, we can repeat the coffee accident on a laboratory benchtop. Paper is an inhomogeneous material, a prototype of the porous inhomogeneous rock that holds oil! One difference between fluid flow in the paper towel and in oil-bearing rock arises from the length scales at which these phenomena take place. This difference is an advantage: we can use a 20 cm paper towel system (Fig. 1.2) to help develop our understanding of the 20 km oilfield problem. For example, we can characterize the wet–dry interface using scaling laws, whose form is predicted by simple models that capture the essential mechanisms contributing to the morphology. This 'benchtop exercise' is an example of some of the current experiments being carried out on idealized systems which are yielding new insights into practical interface problems.

1.1.2 Propagation of flame fronts

Take a sheet of paper and ignite it at one end. Try to keep it horizontal, so that the entire paper does not take flame at once – if possible, use paper that burns without flame. After burning a part of it, inspect the interface between the burned and unburned parts (see Fig. 1.3). Is it rough, similar to the edge of the coffee droplet on the tablecloth? Is this similarity a coincidence, or there is something in common between these processes?

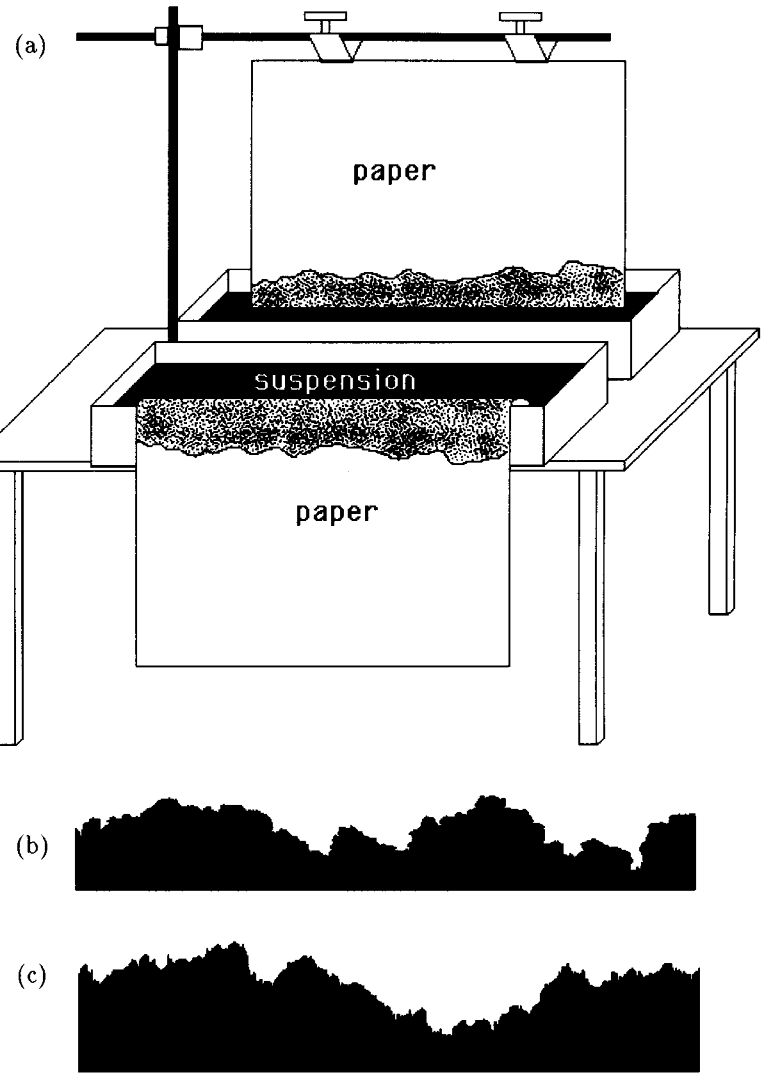


Figure 1.2 (a) Schematic illustration of an experimental setup probing interface motion in random media. Parameters such as type of paper, temperature, humidity, direction of growth and concentration of coffee can be varied systematically. These changes affect the area, the speed of wetting, and the global width of the rough surface, but they do not affect the scaling properties of the surface. (b) Digitized *experimental* interface; the horizontal size of the paper was 20 cm. (c) Typical result of a discrete *model* mimicking interface motion in disordered media (see §10.2). (After [31]).

1.1.3 Flux lines in a superconductor

Suppose we place a superconductor in an external magnetic field. The field penetrates the material by generating flux lines or vortices, each carrying an elementary flux (see Fig. 1.4). If there are no impurities in the superconductor and the temperature is low, the flux lines form an array of straight lines. If there are impurities in the superconductor, the flux lines stretch to get close to the impurity sites. These individual flux lines are rough, resembling the surface of the coffee drop, or the fire front. However, there is one important difference between a flux line and a fire or fluid interface: a firefront is a topologically one-dimensional object moving in a two-dimensional plane, while a

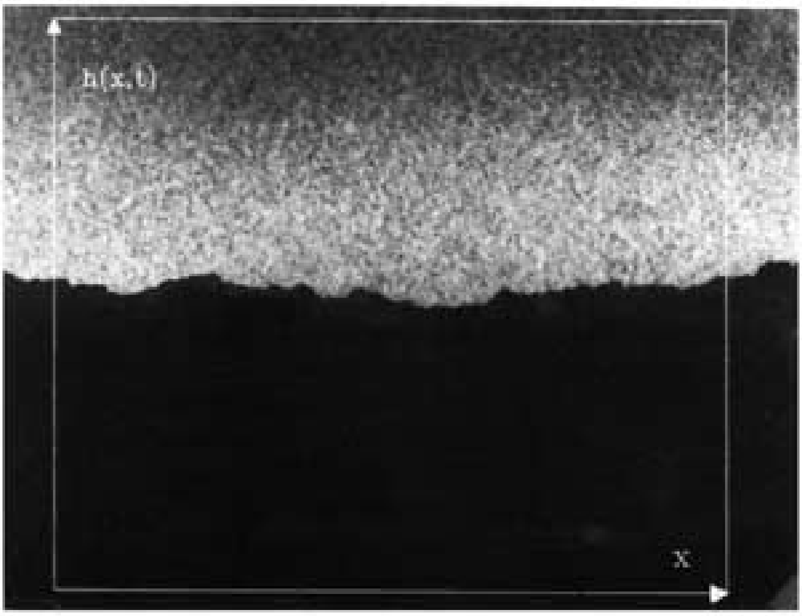


Figure 1.3 An 8 cm segment of paper in which fire propagates from below. (After [504]).

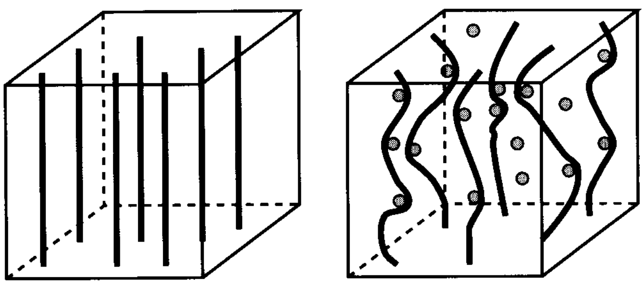


Figure 1.4 Vortex lines in a type II superconductor. (a) In a perfectly clean superconductor at low temperatures the flux lines form an ordered lattice. (b) When the temperature is increased, thermal fluctuations destroy the ordered lattice and the flux lines become wavy. If there are impurities (shaded circles) in the superconductor, the flux lines are pinned in random positions of the impurities.

flux line is a topologically one-dimensional object fluctuating in a *three-dimensional* space.

One purpose of this book is to discuss apparently different phenomena using a common framework. It is coming to be appreciated that all the systems discussed in this section display essentially the same physics: there is an elastic interface ('elastic' in the sense that it does not break, but tries to remain smooth) which propagates in a disordered material. The randomness of the substrate acts to pin the interface, thereby making the interface rough. In the fluid case, local inhomogeneities may block the fluid flow. Some parts of the paper do not burn as rapidly as others, so the flame is halted. Impurities attract the flux line and pin it down in random positions. We shall see that these systems are described by the same laws, and that they can be studied using a similar set of numerical and analytical methods.

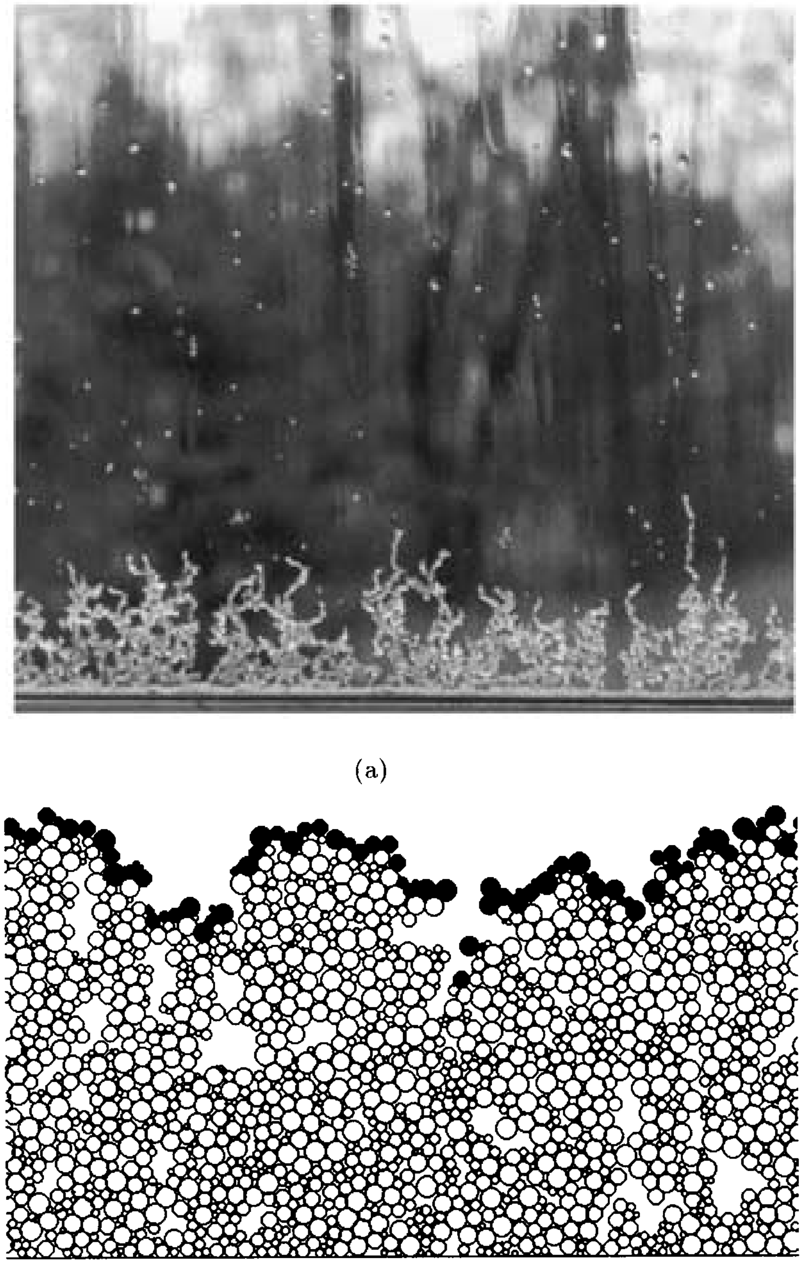
1.2 Deposition processes

Winter. Look out of your car window – snow is falling. The larger snowflakes slide down the window slowly, and form the aggregate shown in Fig. 1.5(a). Notice that at length scales comparable to the size of the snowflake the aggregate is very rough. Why? Snowflakes are deposited randomly. Once they arrive at the aggregate, they stick. The *randomness* in the deposition process apparently leads to a *rough* surface. A similar deposition process is illustrated in Fig. 1.5(b). The resulting bulk is nearly homogeneous, but the surface is quite rough. These are but two of the many examples for which random deposition of some material occurs, and we witness the dynamic growth of a rough surface.

1.2.1 Atom deposition

A deposition process of greater technological importance than snowfall takes place during the growth of thin films by molecular beam epitaxy (MBE), a technology used to manufacture computer chips and other semiconductor devices, indispensable in today's technological world. The most common element used in computer chips is silicon. An example of a very clean Si surface is shown in Fig. 1.6, obtained using a scanning tunneling microscope (STM).

Now imagine that you start depositing new atoms on this Si surface. In contrast to the snowflakes that stick on the first contact point, the Si atom does not stick, but diffuses. When a Si atom reaches the edge of a step, it forms covalent bonds with neighboring atoms, and sticks with a high probability. Such bonds may be broken again, but with a low probability. If the incoming flux is large, there is a large number



(a)

(b)

Figure 1.5 (a) Snow particles falling on a slanted glass window (After [270]); (b) The interface generated by a simple deposition model, in which spherical particles with uniformly distributed random diameters arrive on the surface and roll until they make contact with at least two other discs. (After [89]).

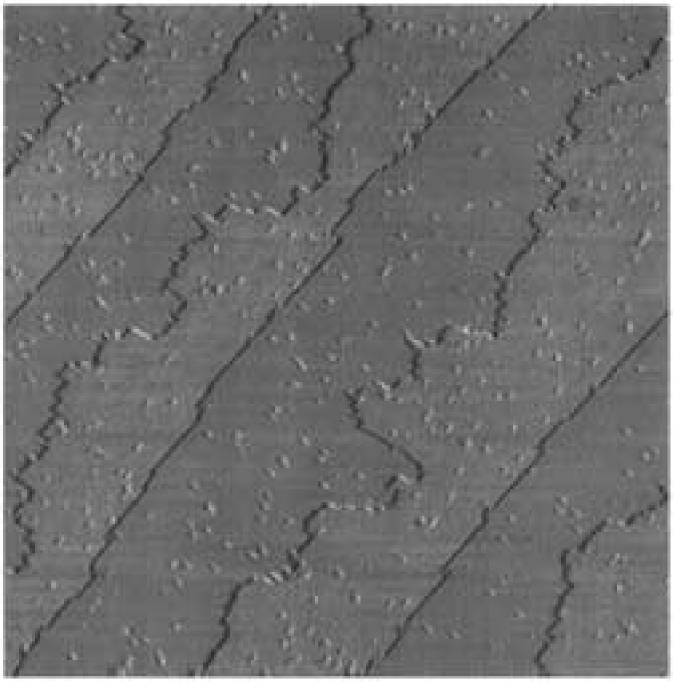


Figure 1.6 A Si surface, as examined by STM. One can distinguish both individual atoms and vacancies. The rugged lines correspond to steps on the surface, where the height of the interface increases by one atom. (Courtesy of M. Lagally).

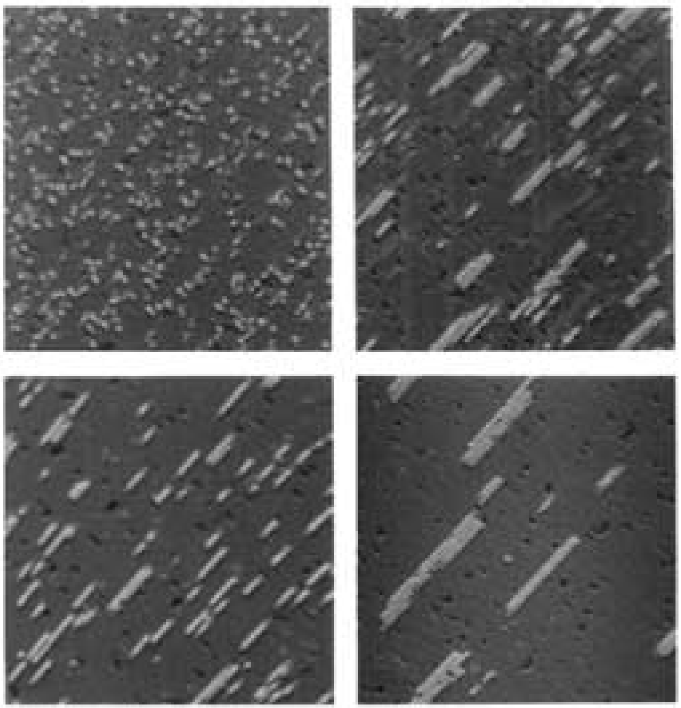
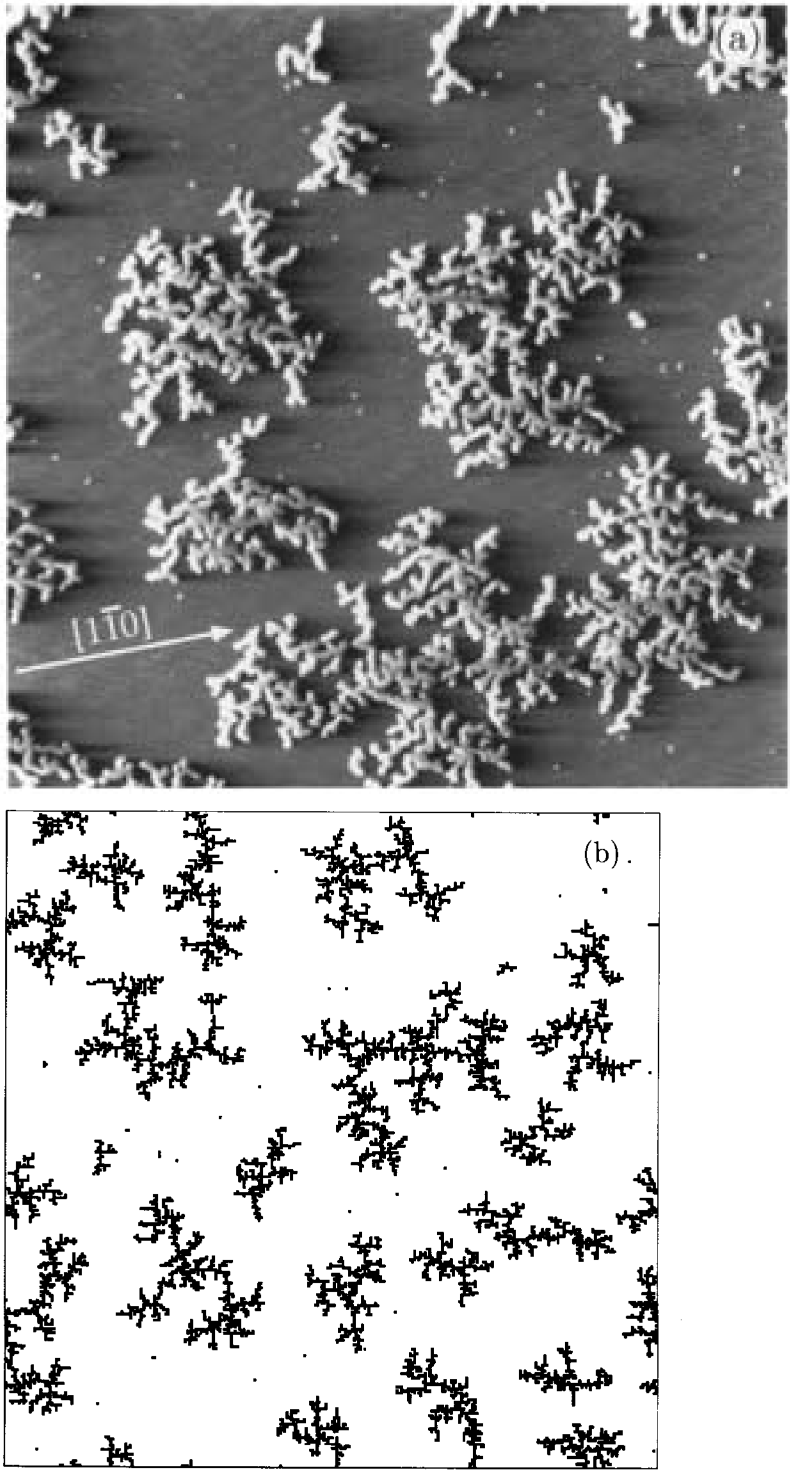


Figure 1.7 Formation of islands by atoms deposition on a Si surface. (Courtesy of M. Lagally).

Figure 1.8
(a) Branched island morphologies obtained by Ag atom deposition on Pt at 110 K. (After [57]). (b) Island formation in a model incorporating the three basic processes taking place during MBE: deposition, diffusion, and aggregation. The deposited atoms aggregate due to diffusion, generating branched fractal islands, similar to those observed in (a). (After [197]).



of wandering atoms on the surface, which meet and 'glue' together, forming islands (see Fig. 1.7).

The islands do not always have a regular shape. As Fig. 1.8 illustrates, Ag deposition on Pt surfaces leads to branched structures, which are known to be self-similar, or fractal. What is the difference between the Si and the Pt that accounts for the difference in island structure? Is it related to the difference in the material properties, or only to the different temperatures and deposition rates?

1.2.2 Roughening in MBE

If we continue to deposit atoms, smaller islands will form on the top of the larger islands. The interface eventually becomes quite rough (see Fig. 1.9). Our discussion so far suggests that we may be happy whenever we see a rough surface. However, engineers usually wish to make a *smooth* film, since rough surfaces have poor contact properties and cannot be used in most applications. In order to avoid roughness, one first must understand the basic mechanism leading to roughness, and the processes affecting the morphology in general. This understanding then may be explored to grow films in regimes where roughening is reduced or absent.

Another common method used in film deposition experiments is sputtering. During *sputter erosion* the material is bombarded with an ion beam that hits the surface and kicks out atoms. This process is used to clean a surface, by eroding a few layers – or, by guiding

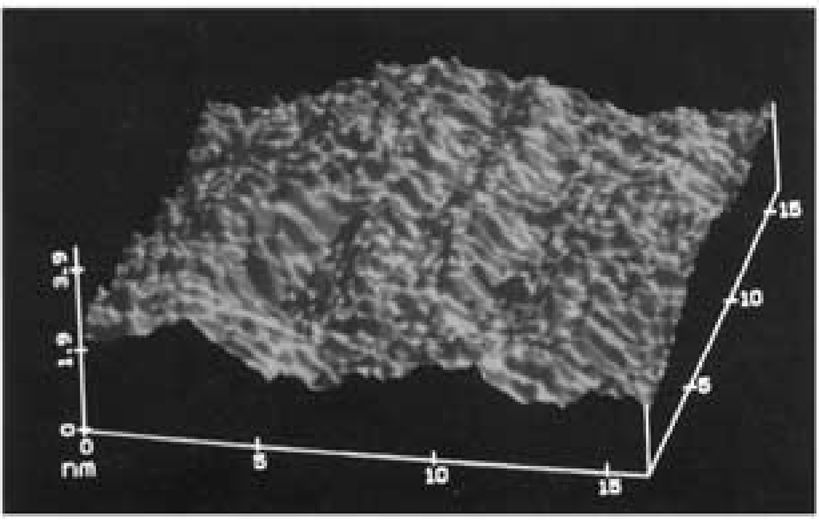


Figure 1.9 STM image of a rough Ag substrate. (After [243]).

the eroded atoms towards a sample, to grow another surface by a process called *sputter deposition*. Films grown by sputter deposition sometimes develop interesting 'cauliflower' structures, as is shown in Fig. 1.10. Why do these manmade films resemble so well the natural cauliflower?

The actual surface morphology in sputter erosion depends on the experimental conditions; some experiments lead to rough interfaces, others to periodic ripple structures (Fig. 1.11). What is the mechanism of the roughening process? Is erosion simply the inverse of deposition, or does it involve processes not present during deposition? How do we explain the formation of the ripple structures on the surface?

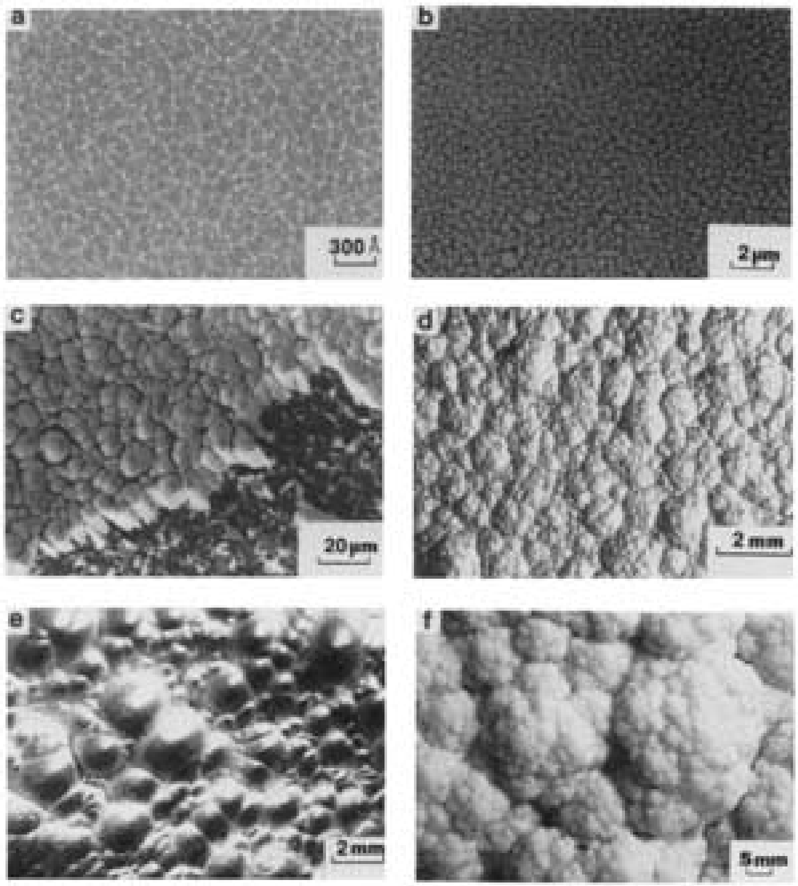


Figure 1.10 Micrographs showing the similarity in morphology for various materials at different magnifications – these span roughly six orders of magnitude in linear dimension. (a) TEM micrograph of *a*-Ge; (b) SEM micrograph of *a*-Si on glass substrate; (c) SEM micrograph of *a*-Si on polycrystalline Al substrate; (d) optical micrograph of pyrolytic graphite; (e) optical micrograph of a thick metal film, and (f) photograph of a cauliflower. (After [316]).

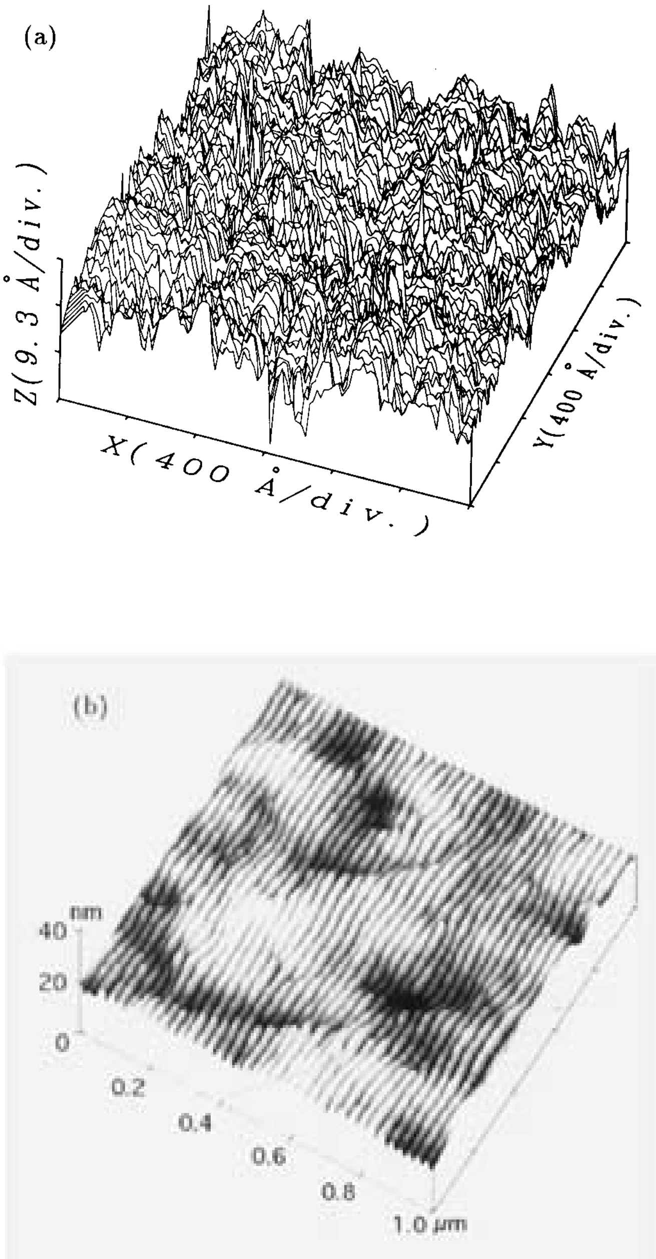


Figure 1.11 (a) STM topograph of a graphite surface after bombarding with a flux $F = 6.9 \times 10^{13}$ ions.cm^{-2}sec^{-1} and an ion fluence $Q = 10^{16}$ ions/cm^2 at room temperature. The sample size is 2400 Å ×2400 Å, and the vertical size 18.6 Å. (After [114]). (b) Atomic force microscope image of a Xe-bombarded SiO_2 film. Note the periodic ripple structure. (After [301]).

1.3 Biological systems

1.3.1 Bacterial growth

The previous examples were selected from the field of materials science. But there are also interesting interfaces in biology. Let us consider a much-studied problem, the growth of bacterial colonies. In a typical experimental setup, agar is prepared in a petri dish. In the middle of the agar a bacterium is inoculated, whereupon it multiplies. At microscopic length scales, the bacteria exhibit a random motion. Looking from a distance, however, a range of interesting morphologies can be observed (Fig. 1.12). The actual morphology depends on the nutrient concentration and on other experimentally-controllable parameters. Some colonies have a compact shape, with a rough surface, similar to the morphology we confronted with spilt coffee. Others are branched, reminiscent of the islands observed in atomic deposition. Are there some general principles common to bacterial growth, island formation, and fluid flow?

1.3.2 DNA

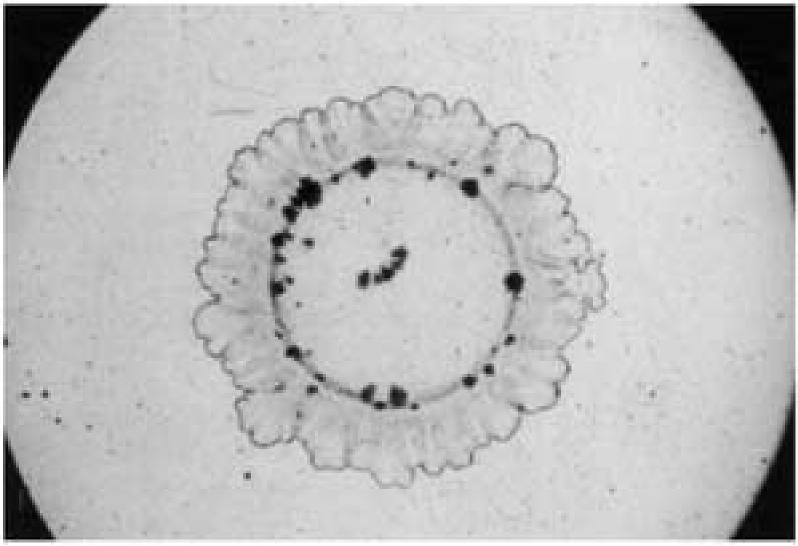
Many systems develop well-defined interfaces. However, there are processes that do not have a surface at all, but with a natural choice of variables can be mapped to a mathematical function or 'landscape' whose roughness is amenable to analysis using the same methods used for real interfaces.

An example that at first sight has nothing to do with interfaces is one-dimensional Brownian motion. Consider a drunk in an elevator of a skyscraper. Let us imagine that the elevator has only two buttons, up and down: the up button takes the elevator one level up, the down button one level down. The drunk punches the buttons randomly. If we plot the position of the elevator as a function of time, we obtain a jagged landscape, called the trail of the random walk, which may be described by many of the same methods used to quantify interface morphology.

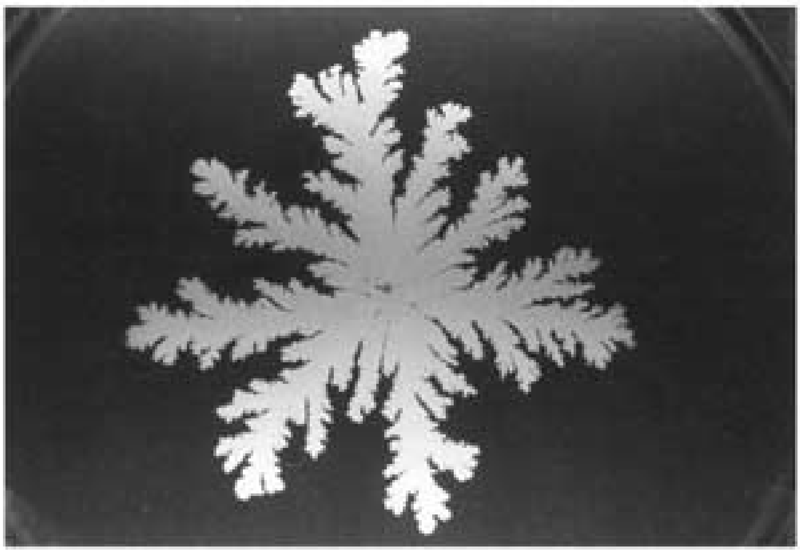
Genomic DNA sequences code for protein structure. The human genome contains information for approximately 100 000 different proteins, which define all inheritable features of an individual – it is likely the most sophisticated information database, created entirely through the dynamic process of evolution.

The building blocks for coding this information, called *base pairs*, form two classes, purines and pyrimidines. In order to study the correlations of a DNA sequence, one can introduce a graphical rep-

Figure 1.12 Examples of bacteria colonies. (a) A colony with roughly compact shape. (After [39].) (b) A colony with a branched morphology, resembling the DLA growth model described in Chapter 19. (After [296]).



(a)



(b)

resentation of the base pair sequence. This 'DNA walk' allows one to visualize directly the fluctuations of the purine-pyrimidine content in DNA sequences: an 'uphill drift' in a region of the landscape corresponds to high local concentration of pyrimidines, while a 'downhill drift' corresponds to high local concentration of purines.

Figure 1.13 shows a typical example of a gene that contains a significant fraction of base pairs that do *not* code for amino acids. The advantage of the DNA walk representation is that it can be quantitatively studied using the methods developed for interfaces. The methods reveal the surprising fact that such noncoding DNA,

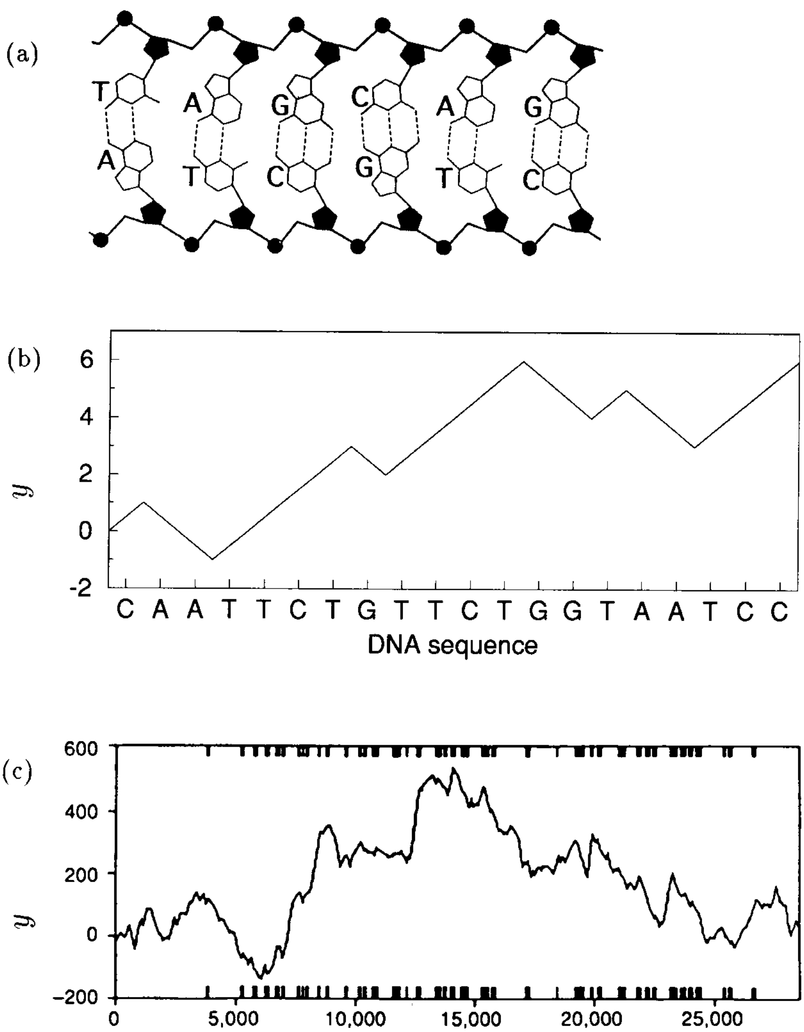


Figure 1.13 (a) The base pairing of two DNA strands to form a double helix. So far as the information content is concerned, a DNA sequence can be represented as a symbolic sequence of four letters: A, C, G and T. (b) Schematic illustration of the definition of the 'DNA walk'. The walker steps 'up' [$u(i) = +1$] if a pyrimidine (C or T) occurs at position i along the DNA chain, while the walker steps 'down' [$u(i) = -1$] if a purine (A or G) occurs at position i. (c) DNA walk for a DNA sequence comprising more than 25 000 base pairs. (After [60]).

previously believed to have at most correlations of very short range, in fact displays long-range correlations [60]. The implications of this result for the possible language characterizing the noncoding DNA is a topic under current investigation [290].

1.4 Methods of analysis

Each field has its own methods for treating a given problem. The development of new approaches, with more descriptive and predicting power, is one of the major goals of science. For the field of disorderly surface growth there are a number of standard tools that must be mastered. We briefly discuss four important methods that are developed and used throughout the book.

1.4.1 Scaling concepts

One of the modern concepts used to study various roughening processes is *scaling*. Scaling has a surprising power of prediction, simple manipulations allowing us to connect apparently independent quantities and exponents.

We shall see that many measurable quantities obey simple *scaling relations*. For example, for a large number of systems we shall find that the interface width, $w(t)$, increases as a power of time, $w(t) \sim t^{\beta}$. The width eventually saturates at a value that increases as a power law of the system size, $w(L) \sim L^{\alpha}$.

Studying such scaling relations will allow us to define *universality* classes. The universality class concept is a product of modern statistical mechanics, and codifies the fact that there are but a few essential factors that determine the exponents characterizing the scaling behavior. Thus different systems which at first sight may appear to have no connection between them, behave in a remarkably similar fashion.

The values of the exponents α and β are independent of many 'details' of the system. For example, α and β do not depend on whether we immerse the paper in ink or coffee, or if we use a paper towel or a tablecloth. In fact, we shall see that the scaling exponents obtained for the fluid flow problem coincide with the scaling exponents obtained for the burning front, despite the rather different mechanisms leading to the actual interface.